Supplemental Document

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1. Radiative cooling background

Radiative cooling structures are designed to selectively emit radiation within the atmospheric transmission window, reflect the solar spectrum, and minimize conductive or convective heating losses. Formally this is expressed as a power balance

$$P_{net} = P_r - P_a - P_{sun} - P_{other} \#(\mathrm{S1})$$

where P_{net} is the net power leaving the structure, P_r is the thermal power the structure emits, P_a is the thermal power emitted from the atmosphere that is absorbed by the radiative cooler, P_{sun} is the solar power absorbed by the radiative cooler, and P_{other} accounts for heating due to conduction or convection. To cool below room temperature, the structure must reflect the solar spectrum to prevent heat buildup and emit within the atmospheric transmission window to radiate its heat into outer space. The cooling power of a radiative cooler is defined by the amount of thermal radiation it emits per unit time and can be expressed as

$$P_r(T) = 2\pi A \int_0^{\pi/2} \int_0^{\infty} I_B(\lambda, T_r) e_r(\lambda, \theta) \sin\theta \cos\theta \, d\lambda d\theta \, \#(\mathrm{S2})$$

where A is the structure area, e_r is the emissivity of the radiative cooler and I_B is the blackbody spectral radiance of the radiative cooler

$$I_B(\lambda, T) = \frac{2hc^2}{\lambda^5} \frac{1}{e^{\frac{hc}{\lambda k_B T}} - 1}, \#(\mathrm{S3})$$

where T_r is the structure's temperature and λ is the emission wavelength.

Under thermodynamic equilibrium, emissivity and absorptivity can be interchanged based on Kirchhoff's law of radiation. Heating of the structure by absorbed atmospheric radiation is expressed as

$$P_a(T) = 2\pi A \int_0^{\pi/2} \int_0^{\infty} I_B(\lambda, T_a) e_r(\lambda, \theta) e_a(\lambda, \theta, \alpha) \sin\theta \cos\theta \, d\lambda d\theta \, \#(\mathrm{S4})$$

where e_a is the emissivity of the atmosphere, $I_B(\lambda, T_a)$ is the blackbody spectral radiance of the atmosphere at ambient temperature T_a, and α is a variable encapsulating the conditions relating to the composition of the atmosphere [S. Jeon and J. Shin, Scientific Reports 10(1), 1-7 (2020)]. The power absorption from direct solar radiation can be expressed as

$$P_{sun} = A\int_0^\infty I_{solar} e_r d\lambda \#(S5)$$

where I_{solar} is the AM1.5 solar spectrum. Finally, heating due to conduction and convection can be collectively expressed as

$$P_{other} = qA(T_a - T_r)\#(S6)$$

where T_a is the ambient temperature, T_r is the temperature of the radiative cooler, and q is the non-radiative heat coefficient from conductive and convective heat transfer through the air and surfaces in contact with the radiative cooler.

Eq. (S1) – Eq. (S6) outline three important facts for radiative cooling structure design. First, the criterion for an optimal cooling structure should be defined by its cooling power at a given operating temperature. This is because as the structure cools below the ambient temperature, the optimal spectral window to achieve maximum cooling power becomes a subset of the atmospheric window. Second, the performance limit for a cooling structure is fundamentally limited by the atmospheric emission spectrum. Third, to achieve net cooling performance, solar absorption and other forms of parasitic heating must be below a critical threshold.

2. Tables of cooling power versus temperature for 2-layer radiative cooling structure design parameters

Table S1. Cooling power versus temperature for Si_3N_4 on SiO_2 (Film/Film) on Ag back reflector

T (K)	P (W/m^2)	Si_3N_4 Thickness (nm)	SiO_2 Thickness (nm)
300	52.39	200	1200
290	30.65	100	1300
280	12.02	100	1100
270	-1.57	600	25

Table S2. Cooling power versus temperature for Si_3N_4 on SiO_2 (NP/Film) on Ag back reflector using the Bruggeman formula (v = 2)

T (K)	P (W/m^2)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)	SiO_2 Thickness (nm)
300	63.76	3000	25	200
290	41.25	2750	25	0
280	22.83	2500	25	0
270	8.01	2500	20	0

Table S3. Cooling power versus temperature for Si_3N_4 on SiO_2 (Film/NP) on Ag back reflector using the Bruggeman formula (v = 2)

T (K)	P (W/m^2)	Si_3N_4 Thickness (nm)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)
300	64.00	200	2500	20
290	37.01	100	2250	30
280	15.82	100	2000	25

270	-0.09	100	1700	20

Table S4. Cooling power versus temperature for Si_3N_4 on SiO_2 (NP/NP) on Ag back reflector using the Bruggeman formula (v = 2)

T (K)	P (W/m^2)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)
300	67.60	1100	35	1800	25
290	42.79	1600	25	1400	20
280	23.10	2500	25	50	20
270	8.10	2500	20	25	20

Table S5. Cooling power versus temperature for SiO_2 on Si_3N_4 (Film/Film) on Ag back reflector

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	Si_3N_4 Thickness (nm)
300	49.57	700	800
290	28.28	800	600
280	10.71	700	600
270	-1.64	0	600

Table S6. Cooling power versus temperature for SiO_2 on Si_3N_4 (NP/Film) on Ag back reflector using the Bruggeman formula (v = 2)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)	Si_3N_4 Thickness (nm)
300	57.78	1700	35	900
290	32.05	1600	35	800
280	11.64	1400	25	700
270	-1.64	0	-	600

Table S7. Cooling power versus temperature for SiO_2 on Si_3N_4 (Film/NP) on Ag back reflector using the Bruggeman formula (v = 2)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)
300	64.91	50	2750	30
290	41.97	25	2750	25
280	22.83	0	2500	25
270	8.01	0	2500	20

Table S8. Cooling power versus temperature for SiO_2 on Si_3N_4 (NP/NP) on Ag back reflector using the Bruggeman formula (v = 2)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)
300	69.43	900	20	2250	35
290	44.38	700	20	2250	30
280	23.58	200	20	2500	25

270	8.01	0	-	2500	20

Table S9. Cooling power versus temperature for Si_3N_4 on SiO_2 (NP/Film) on Ag back reflector using Maxwell Garnett formula (v = 0)

T (K)	P (W/m^2)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)	SiO_2 Thickness (nm)
300	56.82	3000	25	200
290	37.89	2750	25	0
280	21.33	2500	25	0
270	8.38	2500	20	0

Table S10. Cooling power versus temperature for Si_3N_4 on SiO_2 (Film/NP) on Ag back reflector using Maxwell Garnett formula (v = 0)

T (K)	P (W/m^2)	Si_3N_4 Thickness (nm)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)
300	60.94	200	2500	20
290	34.88	100	2250	30
280	15.54	100	2000	25
270	0.59	100	1700	20

Table S11. Cooling power versus optimization temperature for Si_3N_4 on SiO_2 (NP/NP) on Ag back reflector using Maxwell Garnett formula (v = 0)

T (K)	P (W/m^2)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)
300	60.41	1100	35	1800	25
290	38.12	1600	25	1400	20
280	21.95	2500	25	50	20
270	7.22	2500	20	25	20

Table S12. Cooling power versus temperature for SiO_2 on Si_3N_4 (NP/Film) on Ag back reflector using Maxwell Garnett formula (v = 0)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)	Si_3N_4 Thickness (nm)
300	53.47	1700	35	900
290	29.39	1600	35	800
280	10.99	1400	25	700
270	-1.64	0	-	600

Table S13. Cooling power versus temperature for SiO_2 on Si_3N_4 (Film/NP) on Ag back reflector using Maxwell Garnett formula (v = 0)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)

300	58.78	50	2750	30
290	37.49	25	2750	25
280	21.33	0	2500	25
270	6.99	0	2500	20

Table S14. Cooling power versus temperature for SiO_2 on Si_3N_4 (NP/NP) on Ag back reflector using Maxwell Garnett formula (v = 0)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)
300	64.05	900	20	2250	35
290	40.64	700	20	2250	30
280	22.34	200	20	2500	25
270	6.99	0	-	2500	20

Table S15. Cooling power versus temperature for SiO_2 on Si_3N_4 (NP/NP) on Ag back reflector using complementary Maxwell Garnett formula (v' = 0)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)
300	57.17	900	20	2250	35
290	31.51	700	20	2250	30
280	9.18	200	20	2500	25

Table S16. Cooling power versus temperature for SiO_2 on Si_3N_4 (NP/NP) on Ag back reflector using a generalized formula (v = 1)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)
300	69.55	900	20	2250	35
290	44.81	700	20	2250	30
280	24.28	200	20	2500	25

Table S17. Cooling power versus temperature for SiO_2 on Si_3N_4 (NP/NP) on Ag back reflector using Coherent Potential formula (v = 3)

T (K)	P (W/m^2)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)
300	67.18	900	20	2250	35
290	42.11	700	20	2250	30
280	21.42	200	20	2500	25

Table S18. Cooling power at T = 270 K for Si_3N_4 on SiO_2 (NP/NP) on Ag back reflector using different effective medium formulas

v	P (W/m^2)	Si_3N_4 Thickness (nm)	Si_3N_4 Fill Fraction (%)	SiO_2 Thickness (nm)	SiO_2 Fill Fraction (%)
0	7.22	2500	20	25	20
0 (complement)	-5.45	2500	20	25	20

1	8.35	2500	20	25	20
3	6.86	2500	20	25	20

3. Spectral, angular, and polarization resolved emissivity profile of 2-layer radiative cooling structures

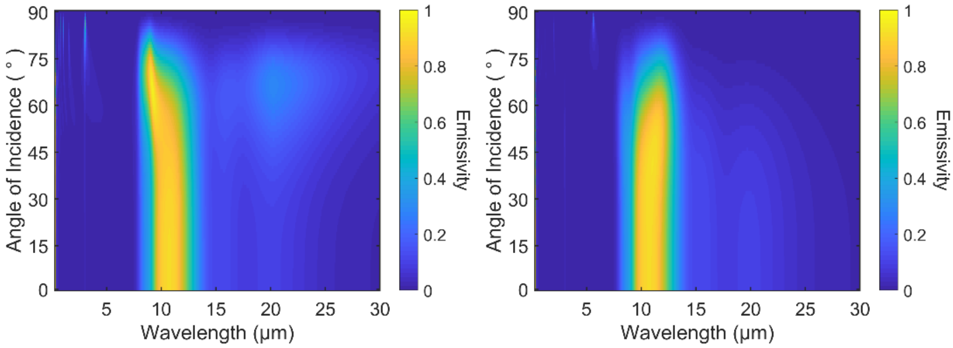

Fig. S1. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer laminate nanoparticle film radiative cooling structure optimized for 270 K at an ambient temperature of 300 K. Radiative cooling structure composed of Si_3N_4 (NP) on SiO_2 (NP) on Ag back reflector.

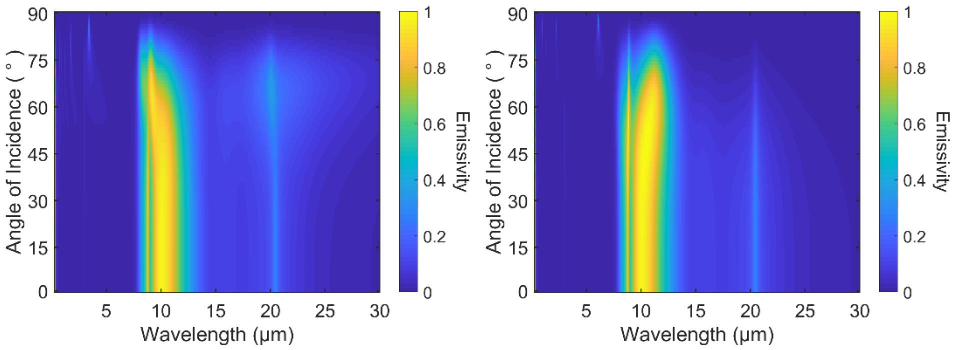

Fig. S2. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer laminate nanoparticle film radiative cooling structure optimized for 280 K at an ambient temperature of 300 K. Radiative cooling structure composed of SiO_2 (NP) on Si_3N_4 (NP) on Ag back reflector.

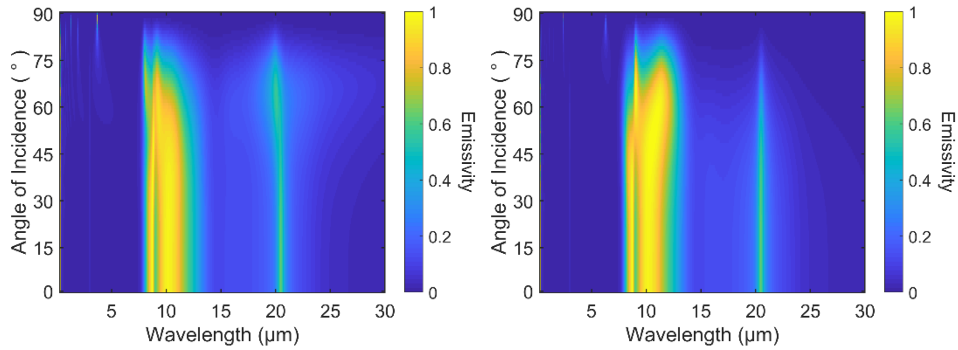

Fig. S3. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer laminate nanoparticle film radiative cooling structure optimized for 290 K at an ambient temperature of 300 K. Radiative cooling structure composed of SiO_2 (NP) on Si_3N_4 (NP) on Ag back reflector.

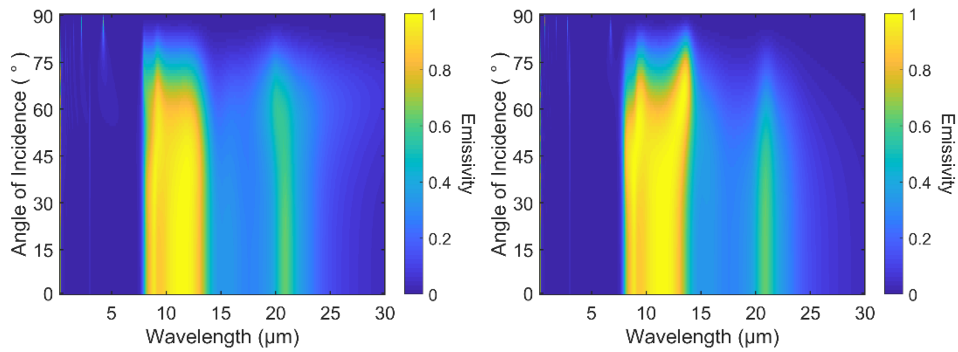

Fig. S4. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer laminate nanoparticle film radiative cooling structure optimized for 300 K at an ambient temperature of 300 K. Radiative cooling structure composed of SiO_2 (NP) on Si_3N_4 (NP) on Ag back reflector.

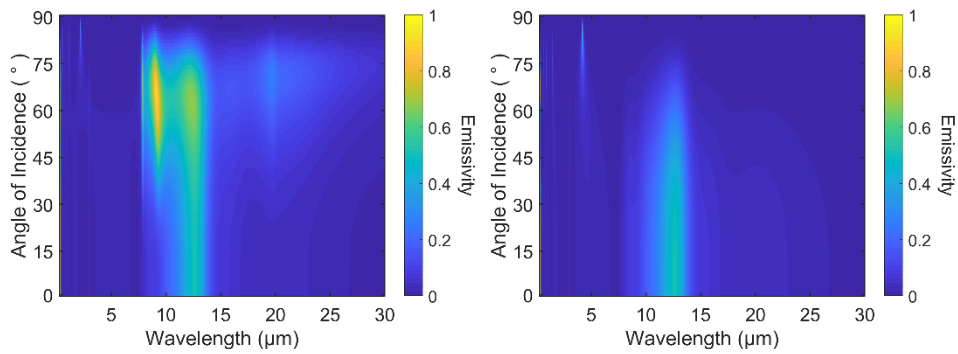

Fig. S5. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer dense solid thin film radiative cooling structure optimized for 270 K at an ambient temperature of 300 K. Radiative cooling structure composed of Si_3N_4 (Film) on SiO_2 (Film) on Ag back reflector.

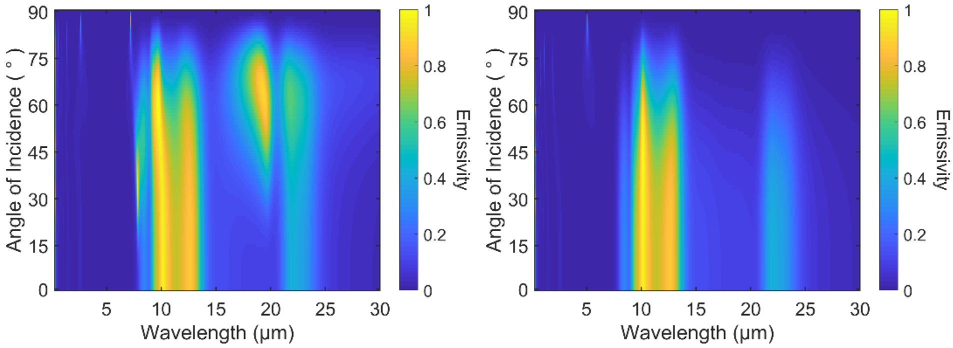

Fig. S6. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer dense solid thin film radiative cooling structure optimized for 280 K at an ambient temperature of 300 K. Radiative cooling structure composed of Si_3N_4 (Film) on SiO_2 (Film) on Ag back reflector.

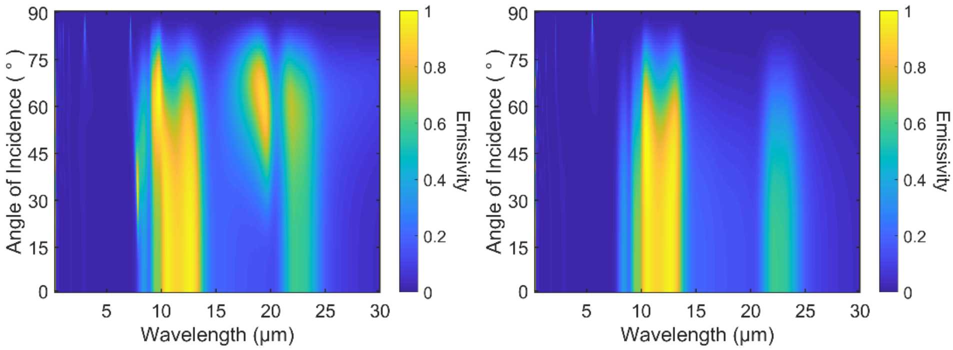

Fig. S7. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer dense solid thin film radiative cooling structure optimized for 290 K at an ambient temperature of 300 K. Radiative cooling structure composed of Si_3N_4 (Film) on SiO_2 (Film) on Ag back reflector.

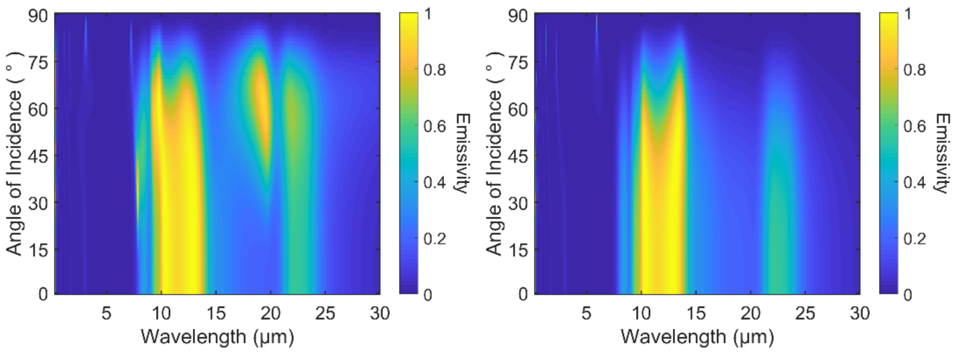

Fig. S8. Spectral and angular resolved p-polarization (left) and s-polarization (right) emissivity profile for 2-layer dense solid thin film radiative cooling structure optimized for 300 K at an ambient temperature of 300 K. Radiative cooling structure composed of Si_3N_4 (Film) on SiO_2 (Film) on Ag back reflector.

4. Numerical emissivity data from various reports in literature

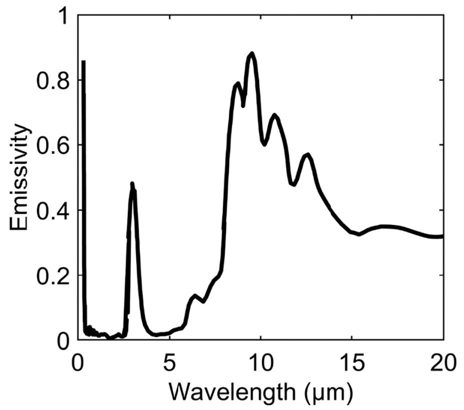

Fig. S9. Digitized emissivity curve from a multilayer radiative cooler composed of seven alternating layers of HfO2 and SiO2. [From Raman *et al.*, Nature **515**(7528), 540 (2014).]

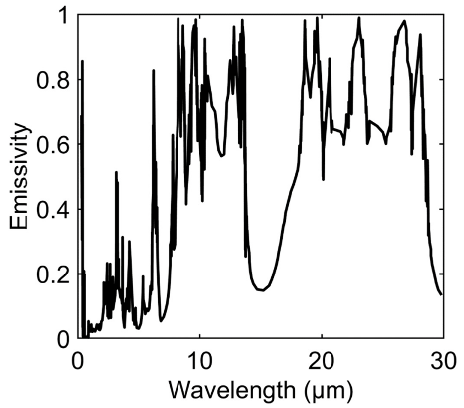

Fig. S10. Digitized emissivity curve from a 2-layer 2D photonic crystal of SiC and quartz. [From Rephaeli *et al.*, Nano Letters **13**(4), 1457-1461 (2013).]

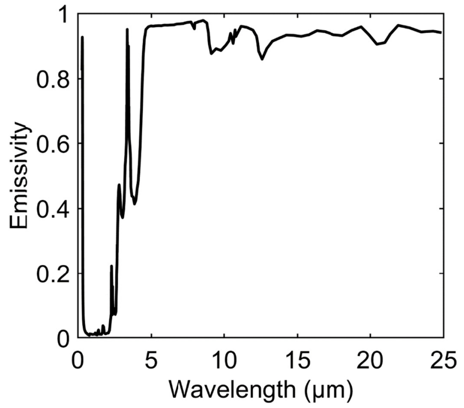

Fig. S11. Digitized emissivity curve from a polymer-coated fused silica mirror. [From Kou *et al.*, ACS Photonics **4**(3), 626-630 (2017).]

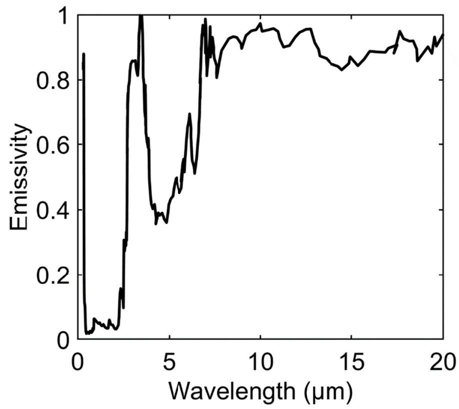

Fig. S12. Digitized emissivity curve from a glass-polymer hybrid metamaterial. [From Zhai *et al.*, Science **355**(6329), 1062-1066 (2017).]

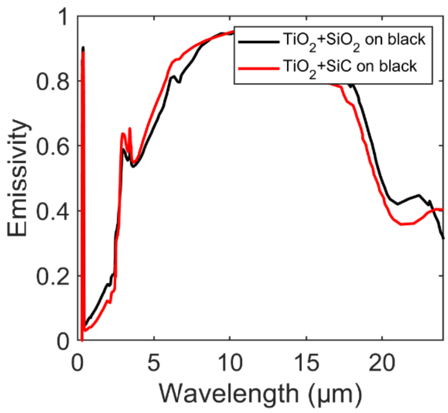

Fig. S13. Digitized emissivity curve from nanoparticle-based double layer cooling structure on a black substrate. [From Bao *et al.*, Solar Energy Materials and Solar Cells **168**, 78-84 (2017).]

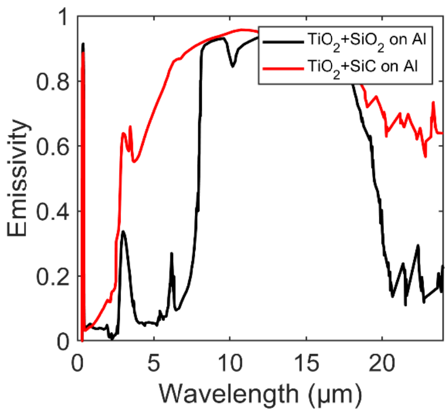

Fig. S14. Digitized emissivity curve from nanoparticle-based double layer cooling structure on an aluminum substrate. [From Bao *et al.*, Solar Energy Materials and Solar Cells **168**, 78-84 (2017).]

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